## **APPENDIX 1**

1. An aromatic fluorophosphorus compound suitable for use as an antioxidant said compound being selected from fluorophosphorus compounds having the structure:

Formula V

wherein R is an substituted aryl group wherein the substituents are tert-alkyl groups:

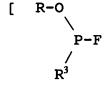
$$(R'O-)_2P-F$$

Formula VI

wherein R' is a substituted aryl group wherein the substituents are selected from sec-alkyl, tertalkyl, aralkyl, cycloalkyl, hydroxy, alkoxy, aryloxy, halo, acyloxy, and alkoxy carbonyl alkyl:]

Formula II

wherein  $R^1$  and  $R^2$  are substituted or unsubstituted [aryl] phenyl groups wherein the [substituent] substituents are selected from alkyl, aryl, aralkyl, cycloalkyl, [hydroxy,] alkoxy, aryloxy, and halo[:], and X is selected from the group consisting of a single bond connecting  $R^1$  and  $R^2$  and divalent bridging groups selected from divalent aliphatic hydrocarbon groups containing 1-12 carbon atoms, -O— and  $-S_q$ — wherein q is an integer from 1 to 3[:], and wherein aryl is selected from the group consisting of phenyl, o-tolyl, p-tolyl, naphthyl, 4-phenylphenyl and 4-sec-hexylphenyl.



Formula III

wherein R is a substituted or unsubstituted aryl group wherein the substituents are selected from alkyl, aryl, aralkyl, cycloalkyl, hydroxy, alkoxy, aryloxy, halo, alkoxycarbonyl, alkoxycarbonyl-alkyl and acyloxy, and R<sup>3</sup> is selected from the group consisting of alkyl, cycloalkyl, aralkyl, aryl, substituted aryl, alkoxy, cycloalkoxy and aralkoxy; and

(HO—) 
$$_{s}$$
—A (—O—P $\stackrel{\checkmark}{\stackrel{}{\sim}}$  R<sup>4</sup>  $_{r}$  Formula IV

wherein A is a mono- or poly-nuclear aromatic group,  $R^4$  is independently selected from fluorine, aryloxy, alkylaryloxy, alkoxy and polyalkoxy, r is an integer from 1 to 4, s is an integer from 0 to 3 and (r + s) equals the valence of A.]

- [2. A compound of claim 1 namely bis(2,6-di-tertbutylphenyl) fluorophosphite.]
- [3. A compound of claim 1 namely: bis(2,4-di-tertbutylphenyl) fluorophosphite.]
- [4. A compound of claim 1 namely bis(4-octadecyloxycarbonylethyl-2,6-di-tert-butylphenyl) fluorophosphite.]
- 5. A compound of claim 1 namely: 2,2'ethylidenebis(4,6-di-tert-butylphenyl) fluorophosphite.
- [6. A compound of claim 1 namely: bis-difluorophosphite ester) of 4,4'-methylenebix(2,6-di-tert-butylphenol).]
- 7. A compound of claim 1 namely: 2,2'-bis(4,6-di-tert-butylphenyl) fluorophosphite.
- 8. Organic material normally susceptible to gradual oxidative degradation when in contact with oxygen, said organic material being a polymer of an olefinically unsaturated monomer and having incorporated therein by mixing or spraying [containing] an antioxidant amount of an aromatic fluorophosphorus compound, said compound being characterized by having at least one benzene group bonded through oxygen to a trivalent phosphorus atom and at least one fluorine atom bonded to said phosphorus atom.
- 9. An organic composition of claim 8 wherein said fluorophosphorus compound is selected from the group consisting of compounds having the structures:

$$(RO-)_n P(-F)_{3-n}$$
 Formula I

wherein R is a substituted or unsubstituted [aryl] <u>phenyl</u> group wherein the substituents are selected from alkyl, aryl, aralkyl, cycloalkyl, [hydroxy,] alkoxy, aryloxy, halo, alkoxycarbonyl,

alkoxycarbonylalkyl and acyloxy and n is 1 or 2, and wherein aryl is selected from the group consisting of phenyl, o-tolyl, p-tolyl, naphthyl, 4-phenylphenyl and 4-sec-hexylphenyl;

wherein R<sup>1</sup> and R<sup>2</sup> are substituted or unsubstituted [aryl] <u>phenyl</u> groups wherein the substituents are selected from alkyl, aryl, aralkyl, cycloalkyl, [hydroxy,] alkoxy, aryloxy and halo, and X is selected [rom] <u>from</u> the group consisting of a single bond connecting R<sup>1</sup> and R<sup>2</sup> and divalent bridging groups selected from divalent aliphatic hydrocarbons containing 1-12 carbon atoms, —O— and —S<sub>q</sub>— wherein q is an integer from 1 to 3[;], and wherein aryl is selected from the group consisting of pheynl, o-tolyl, p-tolyl, naphthyl, 4-phenylphenyl and 4-sec-hexylphenyl; and

wherein R is as previously defined for Formula I and  $[R_3]$   $\underline{R}^3$  is selected from the group consisting of alkyl, cycloalkyl, aralkyl, aryl, substituted aryl, alkoxy, cycloalkoxy, aryloxy and aralkoxy[; and], and wherein aryl is selected from the group consisting of phenyl, o-tolyl, p-tolyl, naphthyl, 4-phenylphenyl and 4-sec-hexylphenyl.

wherein A is a mono or polynuclear aromatic group,  $R^4$  is independently selected from fluorine, aryloxy, alkaryloxy, alkoxy and polyalkoxy and r is an integer from 1 to 4, s is an integer from 0 to 3 and (r+s) equals the valence of A].

- [10. A composition of claim 8 wherein said organic material is a polymer of an olefinically unsaturated monomer.]
- 11. A composition of claim [9] <u>44</u> wherein said organic material is a polymer of an olefinically unsatruated monomer.

- 12. A composition of claim [11] 9 wherein said compound has Formula I[.], and R is a substituted phenyl group.
- 13. A composition of claim 12 wherein n is 2 and said substituents are selected from alkyls having 1-20 carbon atoms, [aryls having 6-12 carbon atoms] phenyl, o-tolyl, p-tolyl, naphthyl, 4-phenylphenyl, 4-sec-hexylphenyl, aralkyls having 7-12 carbon atoms, cycloalkyls having 5-8 carbon atoms, [hydroxy,] alkoxy having 1-12 carbon atoms, aryloxy having 6-12 carbon atoms, halo, [alkoxycarbonylalkyl having 1-20 carbon atoms in its alkoxy moiety and 1-3 carbon atoms in its alkyl moiety, alkoxycarbonyl having 1-20 carbon atoms in its alkoxy moiety] and acyloxy having 1-4 carbon atoms.
- 14. A composition of claim 13 wherein said substituents are selected from alkyl having 1-20 carbon atoms [and alkoxy carbonylalkyl having 1-20 carbon atoms in its alkoxy moiety and 4 1-3 carbon atoms in its alkyl moiety].
- 15. A composition of claim 14 wherein said fluorophosphite compound is bis(2,6-di-tert-butylphenyl) fluorophosphite.
- 16. A composition of claim 14 wherein said fluorophosphite is bis(2,4-di-tert-butylphenyl) fluorophosphite.
- 17. A composition of claim [14] 12 wherein said fluorophosphite compound is bis(4-octadecyloxycarbonylethyl-2,6-di-tert-butylphenyl) fluorophosphite.
  - 18. A composition of claim 12 wherein n is 1.
- 19. A composition of claim 9 wherein said fluorophosphite compound has Formula II wherein said substituents are selected from alkyl having 1-20 carbon atoms, [aryl having 6-12 carbon atoms,] phenyl, o-tolyl, p-tolyl, naphthyl, 4-phenylphenyl, 4-sec-hexylphenyl, aralkyl having 7-12 carbon atoms, cycloalkyl having 5-8 carbon atoms, [hydroxy,] alkoxy having 1-12 carbon [toms] atoms, aryloxy having 6-12 carbon atoms and halo, and X is selected from the group consisting of a single bond connecting  $R^1$  and  $R^2$  and divalent bridging groups selected from divalent aliphatic hydrocarbon groups containing 1-12 carbon atoms, -O-and -S<sub>q</sub>- wherein q is an integer from 1-3.
- 20. A composition of claim 19 wherein said substituent groups are alkyls containing 1-20 carbon atoms.
- 21. A composition of claim 20 wherein said fluorophosphorus compound is 2,2'-ethylidenebis(4,6-di-tert-butylphenyl) fluorophosphite.

- 22. A composition of claim 20 wherein said fluorophosphorus compound is 2,2'-methylenebis (4-methyl-6-tert-butylphenyl) fluorophosphite.
- 23. A composition of claim 20 wherein said fluorophosphite compound is [22,2,] 2,2'-bis(4,6-di-tert-butylphenyl) fluorophosphite.
- 24. A composition of claim 9 wherin said fluorophosphorus compound has Formula III wherein said substituents are selected from alkyls having 1-20 carbon atoms, [aryls having 6-12 carbon atoms,] phenyl, o-tolyl, p-tolyl, naphthyl, 4-phenylphenyl, 4-sechexylphenyl, aralkyls having 7-12 carbon atoms, cycloalkyls having 5-8 carbon atoms, [hydroxy,] alkoxy having 1-12 carbon atoms, aryloxy having 6-12 carbon atoms, halo, alkoxycarbonylalkyl having 1-20 carbon atoms in its alkoxy moiety and 1-3 carbon atoms in its alkyl moiety, alkoxycarbonyl having 1-20 carbon atoms in its alkoxy moiety and acyloxy having 1-4 carbon atoms, and R³ is selected from alkyl having 1-20 carbon atoms, cycloalkyl having 5-8 carbon atoms and aralkyls having 7-12 carbon atoms which are bonded through [oxyqen] oxygen to phosphorus and aryls having 6-12 carbon atoms, alkyl having 1-20 carbon atoms, cycloalkyls having 5-8 carbon atoms and aralkyls having 7-12 carbon atoms which are bonded directly to said phosphorus.
- [25. A composition of claim 9 wherein said fluorophosphorus compound has Formula IV.]
  - [26. A composition of claim 25 whrerein A has a structure selected from:

Structure IV (iii)

$$R^{3} \left( \begin{array}{c} O \\ \parallel \\ -C-C, H_{2}, \end{array} \right)$$

## Structure IV (vii)

wherein  $R^5$  and  $R^6$  are hydrogen or alkyl having 1-12 carbon atoms, y is an integer from 2 to 3, x is an integer from 1 to 3, t is an integer from 2 to 3, u is an integer from 0 to 4 (t+u) equals 2 to 6, w is an integer from 1 to 4,  $R^7$  is hydrogen or an alkyl having 1 to 6 carbon atoms,  $R^8$  is an aliphatic hydrocarbon radical having 1-30 carbon atoms and having valence w, v is an integer from 0-4,  $R^9$  is an aliphatic hydrocarbon radical having 1 to 6 carbon atoms and having valence y.]

- [27. A composition of claim 26 wherein said fluorophosphorus comopund is 2,5-di-tert-butyl-1,4-phenylene bis (difluorophosphite).]
- [28. A composition of claim 26 wherein said fluorophosphorus compound is 4,4'-methylenebis(2,6-di-tert-butylphenyl) bis(difluorophosphite).]
- [29. A composition of claim 26 wherein said fluorophosphite compound is the tris(difluorophosphite ester) of 1,3,5-tris(3,5-di-tert-butyl-4-hydroxybenzyl)-2,4,6-trimethyl benzene.]
- [30. A composition of claim 26 wherein said fluorophosphorus compound is the tetrakis(difluorophosphite ester) of tetrakis(methylene 3-(3,5-di-tert-butyl-4-hydroxyphenyl) propionate)methane.]
- [31. A composition if claim 26 wherein said fluorophosphite compound is difluorophosphite ester of octadecyl 3-(3,5,-di-tert-butylhydroxyphenyl)propioniate.]
- 32. An organic composition of claim 8 further characterized by containing about 0.005-5 wt. percent of a phenolic antioxidant.
- 33. An organic composition of claim 9 further characterized by containing about 0.005-5 wt. percent of a phenolic antioxidant.
- 34. An organic composition of claim 12 further characterized by containing about 0.005-5 wt. percent of a phenolic antioxidant.
- 35. An organic composition of claim 15 further characterized by containing about 0.005-5 wt. percent of a phenolic antioxidant.
- 36. An organic compostion of claim 16 further characterized by containing about 0.005-5 wt. percent of a phenolic antioxidant.

- 37. An organic composition of claim 17 further characterized by containing about 0.005-5 wt. percent of a phenolic antioxidant.
- 38. An organic composition of claim 19 further characterized by containing 0.005-5 wt. percent of a phenolic antioxidant.
- 39. An organic composition of claim 21 further characterized by containing 0.005-5 wt. percent of a phenolic antioxidant.
- 40. An organic composition of claim 39 wherein said phenolic antioxidant is 1,3,5-tris(3,5-di-tert-butyl-b 4-hydroxybenzyl)-2,4,6-trimethylbenzene.
- [41. An organic composition of claim 39 further characterized by containing about 0.005-5 wt. percent of a phenolic antioxidant.]
- [42. An organic composition of claim 25 further characterized by containing about 0.005 -5 wt. percent of a phenolic antioxidant.]
- --43. A aromatic fluorophosphorus compound suitable for use as an antioxidant, said compound being selected from the group consisting of bis(2,4-di-tert-butylphenyl) fluorophosphite; bis(4-octadecyloxycarbonylethyl-2,6-di-tert-butylphenyl) fluorophosphite; and 4,4'-methylenebis(2,6-di-tert-butylphenyl)bis (difluorophosphite).--
- --44. A compound of claim 1 combined in an antioxidant amount with an organic material normally susceptible to gradual oxidative degradation when in contact with oxygen.--

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